

# Spin fluctuations in nearly magnetic metals from ab-initio dynamical spin susceptibility calculations: application to $Pd$ and

$Cr_{95}V_5$

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## Abstract

We describe our theoretical formalism and computational scheme for making *ab-initio* calculations of the dynamic paramagnetic spin susceptibilities of metals and alloys at finite temperatures. Its basis is Time-Dependent Density Functional Theory within an electronic multiple scattering, imaginary time Green function formalism. Results receive a natural interpretation in terms of overdamped oscillator systems making them suitable for incorporation into spin fluctuation theories. For illustration we apply our method to the nearly ferromagnetic metal  $Pd$  and the nearly antiferromagnetic chromium alloy  $Cr_{95}V_5$ . We compare and contrast the spin dynamics of these two metals and in each case identify those fluctuations with relaxation times much longer than typical electronic ‘hopping times’.

75.40Gb, 75.10Lp, 71.15Mb, 75.20En

There is renewed interest in spin fluctuations in materials close to magnetic order. This is due in part to a realisation that nearly critical magnetic fluctuations may be important factors governing the non-conventional properties of a wide range of materials which include the high  $T_c$  superconducting cuprates and heavy fermion systems [1]. The strongly correlated electrons in many of these systems, however, have meant that most theoretical work has concentrated on parameterised models in which the electronic motion is treated rather simply. Another complementary approach is to use an ab initio theory such as Time Dependent Density Functional Theory (TDDFT) [2] but apply it to materials where it can be expected to work i.e. where the effects of electron correlations are not so important, but which otherwise have important similarities to the systems in question. For example, with its perovskite structure containing transition metal(TM)-oxygen planes,  $Sr_2RuO_4$  has several aspects in common with the HTC materials. But the presence of the 4d TM  $Ru$  rather than the narrower band 3d TM  $Cu$  means that electron correlation effects are smaller and therefore DFT-based calculations can provide a valuable starting point. Moreover, its exotic superconductivity at low temperature seems likely to be affected by spin fluctuations [3–5]. Concerning another example, the transition temperature separating paramagnetic and magnetically ordered phases of the cubic transition metal compound  $MnSi$ , which has the B20 crystal structure, is driven down to zero temperature upon the application of pressure. In the vicinity of the critical pressure for this quantum phase transition the system exhibits non- or marginal Fermi liquid properties [6]. In this paper we investigate the energy and temperature dependence of spin fluctuations in two systems which although structurally and compositionally simpler than the two mentioned above, are also close to magnetic phase transitions. The first is palladium which is nearly ferromagnetic and the second is the nearly anti-ferromagnetic  $Cr_{95}V_5$  alloy.

Theoretical models in which an effective action for the slow spin fluctuations is written down have contributed greatly to our understanding of the properties of itinerant electron systems close to magnetic order [6,7]. Recent work which can incorporate results from DFT-based ‘Fixed Spin Moment’ (FSM) electronic structure calculations, treats these fluc-

tuations classically [8]. A Landau-Ginzburg-like energy functional is written down, a free energy constructed which includes terms describing the interactions between the fluctuations (the mode-mode coupling) and properties such as the static susceptibility, specific heat and resistivity calculated. The FSM electronic structure calculations can be used to determine the coefficients in this functional [8]. Many informative studies have been carried out. For those calculations with such a DFT basis, these still remain qualitative investigations because of the lack of a prescription for the effective number of modes to include in the theory and its variation with temperature [6,7]. Stoner single-particle excitation effects are also largely ignored. Both these issues can be addressed by the development and application of methods to calculate the temperature-dependent, dynamic paramagnetic spin susceptibility of nearly magnetic materials. The development of dynamic susceptibility calculations is particularly pertinent now that inelastic neutron scattering experiments, such as the time of flight measurements, have developed to the extent that spin fluctuations in nearly magnetic metals can be accurately measured [9].

We have recently devised and proven a new scheme for calculating the wave-vector and frequency dependent dynamic spin susceptibility of metallic systems [10] which is based on the Time Dependent Density Functional Theory (TDDFT) of Gross et al. [2] and as such is an all electron theory. This enables the temperature dependent dynamic spin susceptibility of metals and compositionally disordered alloys to be calculated.

Over the past few years great progress has been made in establishing TDDFT [2]. Analogs of the Hohenberg-Kohn [11] theorems of the static density functional (DFT) formalism have been proved and rigorous properties found. By considering a paramagnetic metal subjected to a small, time-dependent external magnetic field,  $\mathbf{b}(\mathbf{r}, t)$  which induces a magnetisation  $\mathbf{m}(\mathbf{r}, t)$  and using TDDFT in [10] an expression for the dynamic paramagnetic spin susceptibility  $\chi(\mathbf{q}, w)$  via a variational linear response approach can be derived [12]. Accurate calculations of dynamic susceptibilities from this basis have been scarce (e.g. [13]) because they are difficult and computationally demanding. In ref. [10] we showed that these problems can be mitigated by accessing  $\chi(\mathbf{q}, w)$  via the corresponding *temperature* susceptibility

$\bar{\chi}(\mathbf{q}, w_n)$  where  $w_n$  denotes a bosonic Matsubara frequency [14]. We demonstrated this approach by an investigation into the nature of the spin fluctuations in paramagnetic  $Cr$  and compositionally disordered  $Cr_{95}V_5$  and  $Cr_{95}Re_5$  alloys with good agreement with experimental data [10]. For example, recent inelastic neutron scattering experiments [9,15] have measured incommensurate AF ‘paramagnons’, persisting up to high frequencies in the latter system which were also shown in our calculations

In [10] we provided a brief outline of this approach for crystalline systems with one atom per unit cell only so in the next section we provide its generalisation to multi-atom per unit cell materials in which one or more sublattices may be substitutionally disordered. We also describe the techniques involved in the method in more detail. In the following section we discuss our study of the frequency, wave-vector and temperature dependence of the spin fluctuations in  $Pd$  and identify the paramagnon regime. We then move on to carry out a similar study of the incommensurate antiferromagnetic spin fluctuations in the dilute  $Cr_{95}V_5$  alloy. In each case the results can be interpreted simply in terms of an over-damped harmonic oscillator model [6]. In the latter case we show how the parameters scale with temperature and compare and contrast the two systems. In the final section we draw some conclusions and remark how this work has the potential to be integrated into spin fluctuational theories of nearly and weakly magnetic metals [6].

## I. THE DYNAMICAL SPIN SUSCEPTIBILITY $\chi(\mathbf{Q}, W)$ .

The equilibrium state of a paramagnetic metal, described by standard DFT, has density  $\rho_0(\mathbf{r})$  and its magnetic response function

$$\chi(\mathbf{r}t; \mathbf{r}'t') = \frac{\delta m[b](\mathbf{r}, t)}{\delta b(\mathbf{r}', t')} \Big|_{b=0, \rho_0} \quad (1.1)$$

is given by the following Dyson-type equation.

$$\chi(\mathbf{r}t; \mathbf{r}'t') = \chi_s(\mathbf{r}t; \mathbf{r}'t') + \int d\mathbf{r}_1 \int dt_1 \int d\mathbf{r}_2 \int dt_2 \chi_s(\mathbf{r}t; \mathbf{r}_1 t_1) K_{xc}(\mathbf{r}_1 t_1; \mathbf{r}_2 t_2) \chi(\mathbf{r}_2 t_2, \mathbf{r}'t') \quad (1.2)$$

$\chi_s$  is the magnetic response function of the Kohn-Sham non-interacting system with the same unperturbed density  $\rho_0$  as the full interacting electron system, and

$$K_{xc}(\mathbf{r}t; \mathbf{r}'t') = \frac{\delta b_{xc}(\mathbf{r}, t)}{\delta m(\mathbf{r}', t')} \Big|_{b=0, \rho_0} \quad (1.3)$$

is the functional derivative of the effective exchange-correlation magnetic field with respect to the induced magnetisation. As emphasised in ref. [2] eq.1.2 represents an exact representation of the linear magnetic response. The corresponding development for systems at finite temperature in thermal equilibrium has also been described [12]. In practice approximations to  $K_{xc}$  must be made and this work employs the adiabatic local approximation (ALDA) [2] so that

$$\begin{aligned} K_{xc}^{ALDA}(\mathbf{r}t; \mathbf{r}'t') &= \frac{db_{xc}^{LDA}(\rho(\mathbf{r}, t), m(\mathbf{r}, t))}{dm(\mathbf{r}, t)} \Big|_{\rho_0, m=0} \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \\ &= I_{xc}(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \end{aligned} \quad (1.4)$$

On taking the Fourier transform of eq.1.2 with respect to time we obtain the dynamic spin susceptibility  $\chi(\mathbf{r}, \mathbf{r}'; w)$ .

For computational expediency we consider the corresponding *temperature* susceptibility [14]  $\bar{\chi}(\mathbf{r}, \mathbf{r}'; w_n)$  which occurs in the Fourier representation of the temperature function  $\bar{\chi}(\mathbf{r}\tau; \mathbf{r}'\tau')$  that depends on imaginary time variables  $\tau, \tau'$  and  $w_n$  are the bosonic Matsubara frequencies  $w_n = 2n\pi k_B T$ . Now  $\bar{\chi}(\mathbf{r}, \mathbf{r}'; w_n) \equiv \chi(\mathbf{r}, \mathbf{r}'; iw_n)$  and an analytical continuation to the upper side of the real  $w$  axis produces the dynamic susceptibility  $\chi(\mathbf{r}, \mathbf{r}'; w)$ .

We define our general system as having a crystal structure with lattice vectors  $\{\mathbf{R}_i\}$  and where there are  $N_s$  non-equivalent atoms per unit cell. On the  $l^{th}$  of the  $N_s$  sublattices there are  $N_l$  atomic species with concentrations  $c_{\alpha_l}^{(l)}$  ( $\alpha_l = 1, \dots, N_l$ ). In each unit cell the  $N_s$  atoms are situated at locations  $\mathbf{a}_l, l = 1, \dots, N_s$ . The volume of the unit cell is  $V_{WS}$ . On carrying out a lattice Fourier transform over the lattice vectors  $\{\mathbf{R}_i\}$  we obtain the following Dyson equation for the temperature susceptibility

$$\bar{\chi}^{\alpha_l}(\mathbf{x}_l, \mathbf{x}'_{l'}, \mathbf{q}, w_n) = \sum_{\gamma_{l'}}^{N_{l'}} c_{\gamma_{l'}}^{l'} \bar{\chi}_s^{\alpha_l \gamma_{l'}}(\mathbf{x}_l, \mathbf{x}'_{l'}, \mathbf{q}, w_n)$$

$$+ \sum_{l''}^{N_s} \int d\mathbf{x}_{l''}'' \sum_{\gamma_{l''}}^{N_{l''}} c_{\gamma_{l''}}^{l''} \bar{\chi}_s^{\alpha_l \gamma_{l''}}(\mathbf{x}_l, \mathbf{x}_{l''}'', \mathbf{q}, w_n) I_{xc}^{\gamma_{l''}}(\mathbf{x}_{l''}'') \bar{\chi}^{\gamma_{l''}}(\mathbf{x}_{l''}'', \mathbf{x}_{l'}', \mathbf{q}, w_n) \quad (1.5)$$

with  $\mathbf{x}_l, \mathbf{x}_{l'}'$  and  $\mathbf{x}_{l''}''$  measured relative to atomic cells centred on  $\mathbf{a}_l, \mathbf{a}_{l'}$  and  $\mathbf{a}_{l''}$  respectively.  $\bar{\chi}_s^{\alpha_l \gamma_{l''}}$  describes the non-interacting susceptibility *averaged over all configurations* in which one site on sublattice  $l$  is occupied by an  $\alpha_l$  atom and another site on sublattice  $l'$  is occupied by a  $\gamma_{l'}$  atom.  $\bar{\chi}^{\alpha_l}$  is the full susceptibility averaged over all configurations where an  $\alpha_l$  atom is located on one site on sublattice  $l$ .

In terms of the lattice Fourier transform of the DFT Kohn-Sham Green function of the static unperturbed system the non-interacting susceptibility can be written approximately [18]

$$\bar{\chi}_s^{\alpha_l \gamma_{l'}}(\mathbf{x}_l, \mathbf{x}_{l'}', \mathbf{q}, w_n) = -\frac{1}{\beta} \sum_m \int \frac{d\mathbf{k}}{v_{BZ}} \langle G(\mathbf{x}_l, \mathbf{x}_{l'}', \mathbf{k}, \mu + i\nu_m) \rangle_{\alpha_l \gamma_{l'}} \langle G(\mathbf{x}_{l'}', \mathbf{x}_l, \mathbf{k} + \mathbf{q}, \mu + i(\nu_m + w_n)) \rangle_{\gamma_{l'} \alpha_l} \quad (1.6)$$

where the integral is over the Brillouin zone of the lattice and  $\mathbf{k}, \mathbf{q}$  and  $\mathbf{k} + \mathbf{q}$  are all wavevectors within this Brillouin zone which has volume  $v_{BZ}$ .  $\mu$  is the chemical potential and  $\nu_m$  is a fermionic Matsubara frequency  $(2n + 1)\pi k_B T$ .

The Green function can be obtained within the framework of multiple scattering (KKR) theory [16] and this makes this formalism applicable to disordered alloys as well as ordered compounds and elemental metals, the disorder being treated by the Coherent Potential Approximation (CPA) [17]. Then the partially averaged Green function with species  $\alpha_l$  on a site at  $\mathbf{R}_i + \mathbf{a}_l$  on the  $l^{th}$  sublattice and species  $\gamma_{l'}$  on a site at  $\mathbf{R}_j + \mathbf{a}_{l'}$  on sublattice number  $l'$

$$\langle G(\mathbf{R}_i + \mathbf{a}_l + \mathbf{x}_l, \mathbf{R}_j + \mathbf{a}_{l'} + \mathbf{x}_{l'}', z) \rangle_{\alpha_l, \gamma_{l'}} \quad (1.7)$$

can be evaluated in terms of deviations from the Green function of an electron propagating through a lattice of identical potentials determined by the CPA ansatz [18]. The lattice Fourier transform used in equation (1.6) is expressed as

$$\langle G(\mathbf{x}_l, \mathbf{x}_{l'}', \mathbf{k}, z) \rangle_{\alpha_l \gamma_{l'}} = \sum_{L, L'} Z_L^{\alpha_l}(\mathbf{x}_l, z) \langle \tau_{L, L'}^{l, l'}(\mathbf{k}, z) \rangle_{\alpha_l \gamma_{l'}} Z_{L'}^{\gamma_{l'}}(\mathbf{x}_{l'}', z)$$

$$\begin{aligned}
& + \delta_{l,l'} \sum_{L,L'} [\delta_{\alpha_l \gamma_l} (Z_L^{\alpha_l}(\mathbf{x}_l, z) \langle \tau_{L,L'}^{l,l}(z) \rangle_{\alpha_l} Z_{L'}^{\alpha_l}(\mathbf{x}'_l, z) - Z_L^{\alpha_l}(\mathbf{x}_l^<, z) J_L^{\alpha_l}(\mathbf{x}_l^>, z)) \\
& - Z_L^{\alpha_l}(\mathbf{x}_l, z) \langle \tau_{L,L'}^{l,l}(z) \rangle_{\alpha_l \gamma_l} Z_{L'}^{\gamma_l}(\mathbf{x}'_l, z)]
\end{aligned} \tag{1.8}$$

where  $Z_L^{\alpha_l}$  and  $J_L^{\alpha_l}$  are respectively the regular and irregular solutions of the Schrodinger equation in an atomic cell on sublattice  $l$  inhabited by an  $\alpha_l$  atom and  $L, L'$  represent the angular momentum quantum numbers. The lattice Fourier transform of the averaged scattering path operator  $\tau_{L,L'}^{l,l'}(\mathbf{R}_i - \mathbf{R}_j, z)$  is specified by the following expressions [16]

$$\langle \tau_{L,L'}^{l,l'}(\mathbf{k}, z) \rangle_{\alpha_l \gamma_{l'}} = \sum_{L_1 L_2} D_{\alpha_l, L L_1}^l(z) \bar{\tau}_{L_1, L_2}^{l,l'}(\mathbf{k}, z) \tilde{D}_{\gamma_{l'}, L_2 L'}^{l'} \tag{1.9}$$

where, in matrix notation with respect to angular momentum  $L$ , ( $\tilde{D}$  describes the transpose)

$$D_{\alpha_l, L L_1}^l(z) = [1 + \bar{\tau}^{ll}(z)(m^{\alpha_l}(z) - m^{c,l}(z))]_{L L_1}^{-1} \tag{1.10}$$

with  $m^{\alpha_l}$  being the inverse of the scattering t-matrix  $t^{\alpha_l}$  of an  $\alpha_l$  atom potential,  $m^{c,l}$  the inverse of the CPA t-matrix for the  $l^{th}$  sublattice,  $t^{c,l}$  and  $\bar{\tau}^{ll'}$  the unit cell-diagonal part of the CPA scattering path operator. The following averages also involve these quantities

$$\begin{aligned}
\langle \tau_{L,L'}^{l,l'}(z) \rangle_{\alpha_l \gamma_{l'}} &= \sum_{L_1 L_2} D_{\alpha_l, L L_1}^l(z) \bar{\tau}_{L_1, L_2}^{l,l'}(z) \tilde{D}_{\gamma_{l'}, L_2 L'}^{l'}(z) \\
\langle \tau_{L,L'}^{l,l}(z) \rangle_{\alpha_l} &= \sum_{L_1} D_{\alpha_l, L L_1}^l(z) \bar{\tau}_{L_1, L'}^{l,l}(z)
\end{aligned} \tag{1.11}$$

Finally the lattice Fourier transform of this CPA scattering path operator is given by

$$\bar{\tau}_{L,L'}^{ll'}(\mathbf{k}, z) = t_{L,L'}^{c,l}(z) \delta_{ll'} + \sum_{l''} \sum_{L_1 L_2} t_{L, L_1}^{c,l}(z) g_{L_1 L_2}^{ll''}(\mathbf{k}, z) \bar{\tau}_{L_2 L'}^{l'' l'}(\mathbf{k}, z) \tag{1.12}$$

and  $g_{L_1 L_2}^{ll'}(\mathbf{k}, z)$  are the structure constants for the crystal structure [19].

To solve equation (1.5), we use a direct method of matrix inversion in which, for example,  $\bar{\chi}_s$  is cast into matrix form of order  $(\sum_{l=1}^{N_s} S_l N_l) \times (\sum_{l=1}^{N_s} S_l N_l)$  where  $S_l$  is the number of spatial grid points associated with the  $l^{th}$  sublattice. Local field effects are thus fully incorporated. The full Fourier transform

$$\bar{\chi}(\mathbf{q}, \mathbf{q}; w_n) = (1/V_{WS}) \sum_l \sum_{\alpha_l} c_{\alpha_l}^l \sum_{l'} e^{i\mathbf{q} \cdot (\mathbf{a}_l - \mathbf{a}_{l'})} \int d\mathbf{x}_l \int d\mathbf{x}'_{l'} e^{i\mathbf{q} \cdot (\mathbf{x}_l - \mathbf{x}'_{l'})} \bar{\chi}^{\alpha_l}(\mathbf{x}_l, \mathbf{x}'_{l'}, \mathbf{q}, w_n) \tag{1.13}$$

can then be constructed.

The most computationally demanding parts of the calculation are the convolution integrals over the Brillouin Zone which result from the expression for  $\bar{\chi}_s$ , eq. (1.6) i.e.

$$\int d\mathbf{k} \bar{\tau}_{LL_1}^{ll'}(\mathbf{k}, z) \bar{\tau}_{L_2L_3}^{l'l}(\mathbf{k} + \mathbf{q}, z') \quad (1.14)$$

Since all electronic structure quantities are evaluated at complex energies  $z$ , these convolution integrals have no sharp structure and can be evaluated straightforwardly by an application of the adaptive grid method of B.Ginatempo and E.Bruno [20] which has been found to be very efficient and accurate. In this method one can preset the level of accuracy of the integration by supplying an error parameter  $\epsilon$ . In the calculations described in this paper we have used  $\epsilon = 10^{-4}$  so that we obtain 4 significant figure accuracy on  $\chi_s$ . This control on accuracy is crucial for the description of the long wavelength paramagnons in *Pd*. In this case we needed to sample around 2000  $\mathbf{k}$ -points at energies on the contour distant 0.5Ry. off the real axis but up to 50000  $\mathbf{k}$ -points very close (0.001 Ry.) to the real energy axis. Around half that number of points were required for the *Cr*<sub>95</sub>*V*<sub>5</sub> calculations.

We evaluate the Matsubara sum shown in equation (1.6) by using suitable complex energy contours to enclose some of the Matsubara poles and exploiting the hermiticity of the single-electron Green function  $G(\mathbf{r}, \mathbf{r}', z)$ . Consequently a quantity like  $\chi_s$  at a positive bosonic Matsubara frequency  $w_n$  with form

$$A(\mathbf{r}, \mathbf{r}', w_n) = -\frac{1}{\beta} \sum_m G(\mathbf{r}, \mathbf{r}', \mu + i\nu_m) G(\mathbf{r}', \mathbf{r}, \mu + i\nu_m + iw_n) \quad (1.15)$$

where the fermionic Matsubara sum is over  $m = 0, \pm 1, \pm 2, \dots, \pm \infty$  can be rewritten as

$$A(\mathbf{r}, \mathbf{r}', w_n) = -\frac{2}{\beta} \sum_{m=0}^{M_{max}} \text{Re}(G(\mathbf{r}, \mathbf{r}', \mu + i\nu_m) G(\mathbf{r}', \mathbf{r}, \mu + i\nu_m + iw_n)) + \quad (1.16)$$

$$\frac{1}{\pi} \text{Im} \int_C dz (G(\mathbf{r}, \mathbf{r}', z) G(\mathbf{r}', \mathbf{r}, z + iw_n)) f(z) - \frac{1}{\beta} \sum_{m=0}^{n-1} G(\mathbf{r}, \mathbf{r}', \mu - i\nu_m) G(\mathbf{r}', \mathbf{r}, \mu + iw_n - i\nu_m)$$

Both sums are over positive fermionic frequencies only and  $f(z)$  is the Fermi function.  $C$  is a contour which encloses the first  $M_{max}$  fermionic Matsubara poles with the real axis. In our calculations we have used a simple box contour whose top lies exactly halfway between two

neighbouring  $\nu_m$ 's which makes the Fermi factor at this  $z$  real. The terms in the Matsubara sums for  $\nu_m$ 's  $> 0.001$  are evaluated by interpolating from a grid  $\nu = 0.001 \times 10^{0.1p}$ , with  $p = 0, 1, \dots, 30$  or are evaluated directly.  $M_{max}$  is chosen so  $\nu_{M_{max}} \approx 1$ .

Once the temperature susceptibility  $\bar{\chi}(\mathbf{q}, \mathbf{q}; w_n)$  has been calculated the dynamic susceptibility can be found. As discussed in ref. [14], for example, we can define the retarded response function  $\chi(\mathbf{q}, \mathbf{q}, z)$  of a complex variable  $z$ . Since it can be shown [14] formally that  $\lim_{z \rightarrow \infty} \chi(z) \sim 1/z^2$  and we can obtain  $\chi(iw_n)$  from the above analysis it is possible to continue analytically to values of  $z$  just above the real axis, i.e.  $z = w + i\eta$ . In order to achieve this we fit our data to a function describing a set of overdamped oscillators, i.e.,

$$\bar{\chi}(\mathbf{q}, \mathbf{q}, w_n) = \sum_M \frac{\chi_M(\mathbf{q})}{(1 + (w_n/\Gamma_M(\mathbf{q})) + (w_n/\Omega_M(\mathbf{q}))^2)} \quad (1.17)$$

in which  $M$  is an integer. From this the dynamic susceptibility can be written down and the imaginary part becomes

$$Im\chi(\mathbf{q}, \mathbf{q}, w) = \sum_M \frac{\chi_M(\mathbf{q})(w/\Gamma_M(\mathbf{q}))}{\{[1 - (w/\Omega_M(\mathbf{q}))^2]^2 + (w/\Gamma_M(\mathbf{q}))^2\}}. \quad (1.18)$$

This form ensures that the sum rule involving the static susceptibility  $\chi(\mathbf{q})$  is satisfied, i.e.

$$\chi(\mathbf{q}) = \frac{2}{\pi} \int_0^\infty dw \frac{Im\chi(\mathbf{q}, \mathbf{q}, w)}{w} \quad (1.19)$$

In nearly magnetic metals the nature of the spin fluctuations can be succinctly described via the variance  $\langle m^2(\mathbf{q}) \rangle$ . From the fluctuation dissipation theorem

$$\langle m^2(\mathbf{q}) \rangle = \frac{1}{\pi} \int_{-\infty}^\infty dw \frac{Im\chi(\mathbf{q}, \mathbf{q}, w)}{(1 - e^{-\beta w})} \quad (1.20)$$

By introducing an energy cutoff in this expression the slower fluctuations can be revealed.

We find that very good fits are obtained with small  $M$  in equation (1.17) for a wide range of  $w_n$ 's ( $< 500$  meV) for the systems we have examined to date. For  $Cr$  and its dilute alloys using  $M = 1$  in equation (1.17) provides an excellent fit. This means that a close analogy with an overdamped harmonic oscillator can be made. In the case of palladium, equation (1.17) with  $M = 2$  gives an excellent representation of  $\bar{\chi}(\mathbf{q}, \mathbf{q}, w_n)$ . Now the spin fluctuations can be described in terms of two overdamped oscillators with two distinct timescales. The slower one describes the famous paramagnons of palladium.

## II. THE SPIN FLUCTUATIONS OF NEARLY FERROMAGNETIC PALLADIUM.

Although possessing one of the highest Fermi energy densities of states of all the transition metals, palladium has neither a magnetically ordered nor a superconducting ground state. With its nearly filled d-bands, it is, however, on the verge of being a ferromagnet. This is shown by its uniform paramagnetic spin susceptibility which is greatly enhanced over that of an equivalent non-interacting system by exchange-correlation effects. Moreover, the temperature dependence of the susceptibility also demonstrates effects of long-lived ferromagnetic spin-fluctuations or paramagnons. It has even been proposed that these spin fluctuations might induce p-wave superconductivity in this material on account of similarities with spin-fluctuation induced superfluidity in  $He^3$  [21].

Several theoretical studies of the spin fluctuations of  $Pd$  have been carried out. The early ones were based on a jellium model for the electrons and examined the contribution that spin fluctuations make to the specific heat and their influence on any superconducting phase transition as well as describing the cross-section that would be measured in an inelastic neutron scattering experiment [22]. Later work has attempted to remove the deficiencies of such a single-band model and to describe the band structure effects more realistically [13]. From his calculations of the inelastic neutron scattering cross-section for a range of scattering angles and incident energies, Doniach [22] estimated the circumstances under which the paramagnons of  $Pd$  might be observed. Very recently time of flight measurements on a neutron spallation source have succeeded in detecting these long-wavelength, long-lived spin fluctuations [9].

Using the formalism described in the last section, we have calculated the temperature susceptibility of  $Pd$  at  $T = 100K$  and have then determined the dynamic susceptibility  $\chi(\mathbf{q}, w)$ . Currently our calculations use descriptions of the equilibrium states in terms of scalar-relativistic atomic sphere approximation (ASA) effective one-electron scattering potentials and charge densities. The formalism, however, is applicable to full potentials and non-spherically symmetric charge densities. The lattice spacing of  $3.83 \text{ \AA}$  which we used in

the *Pd* calculations was determined from the minimum of the ASA total energy and found to be 1.5% smaller than the experimentally measured value. At  $15.7 \mu_B^2 eV^{-1}$  our calculated uniform susceptibility  $\chi(0, 0)$  is somewhat lower than the value  $24.3 \mu_B^2 eV^{-1}$  measured experimentally [9]. For small wavevectors  $\mathbf{q}$  the inverse of our calculated static susceptibility depends on the magnitude squared of  $\mathbf{q}$ , i.e.  $\chi^{-1}(\mathbf{q}, 0) \approx \chi^{-1}(0, 0) + cq^2$  as also found experimentally and in other calculations [13]. We obtain a value of approximately  $900 \mu_B^2 \text{\AA}^2 meV$  for  $c$  whereas the value extracted from inelastic neutron scattering experiments is  $294 \pm 130 \mu_B^2 \text{\AA}^2 meV$ . Since palladium is so close to a ferromagnetic phase transition both  $\chi(0, 0)$  and  $c$  are very sensitive to the value of the lattice parameter used and the RPA approximation used for the exchange-correlation interactions. Using the experimental lattice spacing in the calculations greatly increases  $\chi(0, 0)$  (to  $23.5 \mu_B^2 eV^{-1}$  close to the value determined from experiment) and both  $\chi(0, 0)$  and  $c$  are likely to be altered if spin fluctuation interaction effects are treated beyond the LDA [7]. Key aspects of the dynamic behaviour turn out not to be so sensitive.

In figure 1 we show the calculated imaginary part of  $\chi(\mathbf{q}, w)$  for a range of frequencies up to 200 meV for selected wavevectors  $\mathbf{q}$  along the  $(1, 0, 0)$  direction. The sharp peaks at low frequencies for  $q < 0.1$  indicate paramagnon-like behaviour. This is illustrated further in figure 2 which shows the magnetic correlations  $\langle m^2(\mathbf{q}) \rangle$  using energy cutoffs of 500meV and 50meV. Once again the paramagnons are clearly visible for a narrow region of small wave-vectors  $\mathbf{q}$ .

We find that the results can be interpreted in terms of two overdamped simple harmonic oscillators each with a characteristic timescale. The longer timescale one encapsulates the paramagnon behaviour.  $Im\chi(\mathbf{q}, \mathbf{q}, w)$  can now be written in the following way

$$Im\chi(\mathbf{q}, \mathbf{q}, w) \approx \frac{\chi_1(\mathbf{q})(w/\Gamma_1(\mathbf{q}))}{(1 + (w/\Gamma_1(\mathbf{q}))^2)} + \frac{\chi_2(\mathbf{q})(w/\Gamma_2(\mathbf{q}))}{(1 + (w/\Gamma_2(\mathbf{q}))^2)} \quad (2.1)$$

Figure 3(a) shows the quantities  $\chi_1(\mathbf{q})$ ,  $\chi_2(\mathbf{q})$  and their sum whereas figure 3(b) shows  $\Gamma_1^{-1}(\mathbf{q})$  and  $\Gamma_2^{-1}(\mathbf{q})$ . Evidently at small  $q$  the slow relaxation process is dominant and  $\hbar/\Gamma_1(\mathbf{q})$  describes the long time taken for a spin density perturbation to relax back to the

equilibrium value. Moreover, we find that the product of  $\chi_1(\mathbf{q})$  with  $\Gamma_1(\mathbf{q})$  varies as  $\gamma_0 q$  for small  $q$ , so that spin density disturbance decays over a time proportional to the wavelength, i.e. exhibits *Landau damping* [6]. We find  $\gamma_0$  to be  $2.13 \mu_B^2 \text{\AA}$  which is in good agreement with the experimentally determined value of  $1.74 \pm 0.8$  [9] and evidently fairly insensitive to the treatment of exchange-correlation effects.

### III. INCOMMENSURATE ANTIFERROMAGNETIC SPIN FLUCTUATIONS IN

$$Cr_{95}V_5.$$

Chromium, an early 3d transition metal, loses its anti-ferromagnetic ground state when electrons are removed as a suitable dopant is added. For example a strongly exchange-enhanced paramagnet exhibiting anti-ferromagnetic paramagnons is formed when just a few atomic percent of chromium are substituted by vanadium. In this section we explore the similarities and differences between these excitations in one such system,  $Cr_{95}V_5$ , and those reported for palladium. We also examine the temperature dependence.

There is an extensive literature on chromium as the archetypal itinerant anti-ferromagnet (AF) and its alloys [15]. It is well known that  $Cr$ 's famous incommensurate spin density wave (SDW) ground state is linked to the nesting wave-vectors  $\mathbf{q}_{nest}$  identified in the Fermi surface. This feature also carries over to its dilute alloys which have a range of AF properties [15] (see figure 4). Indeed the paramagnetic states of some of these alloys have recently acquired a new relevance on account of analogies drawn with the high temperature superconducting cuprates especially  $(La_cSr_{1-c})_2CuO_4$  [23]. Starting with 'parent' materials  $Cr_{95}Mn_5$  or  $Cr_{95}Re_5$  which are simple commensurate AF materials and then lowering the electron concentration by suitable doping causes the Neél temperature,  $T_N$ , to drop rapidly to zero. The paramagnetic metal which forms for dopant concentrations slightly in excess of the critical concentration for this quantum phase transition is characterised by incommensurate paramagnetic spin fluctuations. In [10] we described the spin fluctuations in the paramagnetic state of three representative systems  $Cr_{95}Re_5$ ,  $Cr$  and  $Cr_{95}V_5$ . We tracked

the tendency for the spin fluctuations to become more nearly commensurate with increasing frequency and also with increasing temperature [10] as observed in experimental data. Our estimates of  $T_N$  in  $Cr_{95}Re_5$  and  $Cr$  of 410K and 280K, respectively, were also in fair agreement with the experimental values of 570K and 311K.

Although there have been several simple parameterised models to describe the magnetic properties of  $Cr$  and its alloys [24], these have all concentrated on the approximately nested electron ‘jack’ and slightly larger octahedral hole pieces of the Fermi surface (FS) [15]. These sheets can be seen clearly in figure 4 which shows the Fermi surface of  $Cr_{95}V_5$ . The FS of  $Cr_{95}Re_5$ ’s is close to being perfectly nested and we calculated in [10] its spin fluctuations above  $T_N$  to be nearly commensurate for small frequencies  $w$ . We also looked at  $Cr$  and  $Cr_{95}V_5$ .  $Cr$ ’s and  $Cr_{95}V_5$ ’s Fermi surfaces are progressively worse nested than  $Cr_{95}Re_5$ ’s. In our calculations for these two systems [10] we found the dominant slow spin fluctuations to be incommensurate with wave-vectors equal to the FS nesting vectors  $\mathbf{q}_{nest}$ .  $Cr_{95}V_5$ ’s is shown on figure 4. At best, simple parameterised models only include the effects of all the remaining electrons via an electron reservoir. Whilst finding the obvious similarities from the FS basis between our results and results from such models, we showed that a complete picture is obtained only when an electronic band-filling effect which favours a simple AF ordering at low temperature is also considered.

As for the case of  $Pd$  we found that the spin fluctuations in the paramagnetic state of the three systems are given an accurate description in terms of a overdamped diffusive simple harmonic oscillator model. Here, however, a single channel is sufficient i.e. the susceptibility closely fits the following

$$Im\chi(\mathbf{q}, \mathbf{q}, w) \approx \frac{\chi(\mathbf{q})(w/\Gamma(\mathbf{q}))}{(1 + (w/\Gamma(\mathbf{q}))^2)} \quad (3.1)$$

In this section we concentrate on the exchange-enhanced paramagnet  $Cr_{95}V_5$  and focus in particular on the temperature dependence of the incommensurate spin fluctuations and show how the parameters of the oscillator model scale with temperature.

Figure 1(a) of reference [10] showed  $Im\chi(\mathbf{q}, \mathbf{q}, w)$  for wavevectors  $\mathbf{q}$  along the  $\{0, 0, 1\}$

direction for frequencies up to 500meV. It showed, in agreement with experiment [9], incommensurate AF paramagnons persisting up to high frequencies with intensity comparable to that at low  $w$ . This is in striking contrast to the paramagnons of *Pd*. Figure 5 shows this persistence of the spin fluctuations in  $Cr_{95}V_5$  to high frequencies in a concise way. It shows the magnetic correlations  $\langle m^2(\mathbf{q}) \rangle$  at 100K for the same two energy cutoffs 50 and 500meV as was used to obtain figure 2 for palladium. Unlike those in the nearly ferromagnetic metal, the magnetic correlations grow by more than an order of magnitude for all the wave-vectors shown as the energy cutoff is extended by a similar factor. The variation with temperature of the dynamic susceptibility and the magnetic correlations have also been investigated. Figure 6 shows  $\langle m^2(\mathbf{q}) \rangle$  calculated with a energy cutoff of 500meV for 50K, 300K and 600K. There is a general trend for the weight to transfer towards the commensurate wavevector  $(0,0,1)$  with increasing temperature. For a smaller energy cutoff  $\langle m^2(\mathbf{q}) \rangle$ , integrated over all  $\mathbf{q}$ , decreases with increasing temperature.

Figures 7(a) and (b) show that both the static susceptibility  $\chi(\mathbf{q})$  and the spin relaxation time  $\Gamma^{-1}(\mathbf{q})/\hbar$  also peak at the incommensurate nesting vector  $(0,0,0.9)$  at low temperature and that weight is shifted towards  $(0,0,1)$  at higher temperatures. Figure 7(b) when compared with the analogous figure 3(b) for *Pd* reveals the main difference between the dominant spin fluctuations in the nearly antiferromagnetic  $Cr_{95}V_5$  and the nearly ferromagnetic *Pd*. For the dominant spin fluctuations, the spin relaxation times are some 50 times faster. The small  $\mathbf{Q}$  behavior is different also. ( $\mathbf{Q} = \mathbf{q} - \mathbf{q}_0$ , where  $\mathbf{q}_0 = 0$  for *Pd* and an incommensurate wave-vector for  $Cr_{95}V_5$ .) The product  $\gamma(\mathbf{q})$  of  $\chi(\mathbf{q})$  and  $\Gamma(\mathbf{q})$  tends to a constant for values of  $\mathbf{q}$  close to  $\mathbf{q}_0$  for  $Cr_{95}V_5$  (consistent with ideas that the dynamical critical exponent [6] should be 2 for antiferromagnetic itinerant electron systems) unlike the linear variation of  $\gamma(\mathbf{q})$  with  $|\mathbf{q}|$  for small  $\mathbf{q}$  found in *Pd*. Between 50K and 300K,  $\gamma(\mathbf{q})$  shows a weak variation with temperature where it has values of roughly  $4 \mu_B^2$  and  $6.5 \mu_B^2$  at the incommensurate wave-vector  $\mathbf{q}_0$  and  $\mathbf{q} = (0,0,1)$  respectively. Above 300K,  $\gamma(\mathbf{q})$  loses the trough around  $\mathbf{q}_0$  and by 600K it is roughly constant at  $5.5 \mu_B^2$  for the range of  $\mathbf{q}$  from  $\mathbf{q}_0$  to  $(0,0,1)$ .

## IV. CONCLUSIONS

We have described the framework and method of implementation of our scheme for carrying out ab-initio calculations of the dynamic paramagnetic spin susceptibilities of solids at finite temperatures. The approach is based upon Time-Dependent Spin Density Functional Theory and is applicable to compositionally disordered alloys with multi-atom per unit cell crystal structures. (We note here that this approach may also be adapted to the study of magnetic excitations in magnetically ordered materials.) From an imaginary time multiple scattering Green function formalism an expression for the temperature susceptibility has been derived and the techniques appropriate for its evaluation described. We have shown how the dynamic susceptibility is obtained from this by analytic continuation from Matsubara frequencies in the complex plane to the real frequency axis [25]. This step provides a natural interpretation of the spin dynamics in terms of overdamped oscillator models.

Although ultimately aiming to investigate the spin fluctuations in systems with complex lattice structures such as  $Sr_2RuO_4$  and  $MnSi$  which are of topical interest, here we have compared and contrasted the nearly ferromagnetic transition metal  $Pd$  with the nearly anti-ferromagnetic dilute  $Cr$  alloy,  $Cr_{95}V_5$ . In both cases we have been able to identify ‘slow’ spin fluctuations so that in due course the mode-mode coupling amongst them may be incorporated into spin-fluctuational theories which describe the low temperature properties of these materials [6,8]. For the case of  $Pd$  the spin susceptibility can be interpreted in terms of two oscillator models. One describes spin fluctuations with fairly ‘fast’ relaxation times but the other describes the spin fluctuations for all wave-vectors which have spin relaxation times more than 40 times as slow as the time taken for an itinerant d-electron to hop between lattice sites. Clearly there is a natural time scale separation between these spin fluctuational modes and the electronic degrees of freedom. Consequently spin fluctuational theory can be used with these modes without the need for any wave-vector cutoff. Work is in progress to carry this out. On the other hand  $Cr_{95}V_5$ ’s dynamic spin susceptibility is interpreted in terms of a single oscillator. Here the spin relaxation times are only some 10

times slower than typical d-electron hopping times for modes with  $\mathbf{q}$ 's in a limited region of the Brillouin zone and therefore mode-mode coupling effects may not be so important. This may explain why our calculations [10] of the static susceptibility of this *Cr* alloy (and *Cr* and its other dilute alloys) receive an adequate description in terms of what is essentially an ab-initio Stoner theory.

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## FIGURES

FIG. 1.  $Im\chi(\mathbf{q}, \mathbf{q}, w)$  of  $Pd$  at  $T = 100K$  in units of  $\mu_B^2 eV^{-1}$  for wave-vectors  $\mathbf{q}$  along the  $\{0, 0, 1\}$  direction where  $\mathbf{q}$  is in units of  $\pi/a$  ( $a$  is the lattice spacing). The frequency axis,  $w$ , is marked in meV. The full curve is for  $\mathbf{q} = (0, 0, 0.02)$ , then for curves with decreasing dash-length,  $\mathbf{q} = (0, 0, 0.1), (0, 0, 0.2)$  and  $(0, 0, 0.5)$ .

FIG. 2. The variance of the spin fluctuations in  $Pd$  at  $T = 100K$ ,  $\langle m^2(\mathbf{q}) \rangle$ , in  $\mu_B^2$  for wave-vectors  $\mathbf{q}$  along the  $\{0, 0, 1\}$  direction with energy cutoffs of 500 meV (full line) and 50 meV (dashed line).

FIG. 3. (a) The static susceptibility of  $Pd$  at  $T = 100K$  partitioned into 2 parts  $\chi_1(\mathbf{q})$ ,  $\chi_2(\mathbf{q})$  of equation (2.1) in units of  $\mu_B^2 eV^{-1}$  for wave-vectors  $\mathbf{q}$  along the  $\{0, 0, 1\}$  direction.  $\chi_1(\mathbf{q}) + \chi_2(\mathbf{q})$  is shown by the full line,  $\chi_1(\mathbf{q})$  by the long-dashed line and  $\chi_2(\mathbf{q})$  by the short-dashed line. (b) The time scales from eq. (2.1) (divided by  $\hbar$ ) in  $eV^{-1}$ ,  $\Gamma_1^{-1}(\mathbf{q})$  (full line),  $\Gamma_2^{-1}(\mathbf{q})$  (dashed line) for  $Pd$  at  $T = 100K$ . Note  $\Gamma_2^{-1}(\mathbf{q})$  has been scaled up by 100 for display purposes.

FIG. 4. The Fermi surface (FS) of  $Cr_{95}V_5$  from Bloch spectral function calculations in the  $q_z = 0$  plane. The dashed arrows mark the nesting vector  $\mathbf{q}_{nest}$  connecting the  $H$ -centered hole octahedron to the slightly smaller  $\Gamma$ -centered electron surface. The FS is well defined with only a small disorder broadening  $< 0.01\pi/a$ .

FIG. 5. The variance of the spin fluctuations in  $Cr_{95}V_5$  at  $T = 100K$ ,  $\langle m^2(\mathbf{q}) \rangle$ , in  $\mu_B^2$  for wave-vectors  $\mathbf{q}$  along the  $\{0, 0, 1\}$  direction with energy cutoffs of 500 meV (full line) and 50 meV (dashed line).

FIG. 6. The variance of the spin fluctuations in  $Cr_{95}V_5$ ,  $\langle m^2(\mathbf{q}) \rangle$ , at  $T = 50K$  (full line), 300K (long-dashed) and 600K (short-dashed), in  $\mu_B^2$  for wave-vectors  $\mathbf{q}$  along the  $\{0, 0, 1\}$  direction with a energy cutoff of 500 meV.

FIG. 7. (a) The static susceptibility of  $Cr_{95}V_5$  at  $T = 50\text{K}$  (full line),  $300\text{K}$  (long-dashed) and  $600\text{K}$  (short-dashed), in  $\mu_B^2 eV^{-1}$  along the  $\{0, 0, 1\}$  direction. (b) The time scale (divided by  $\hbar$ ) in  $eV^{-1}$ ,  $\Gamma^{-1}(\mathbf{q})$  for  $Cr_{95}V_5$  at the same temperatures.

















